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Circular economy could expose children to hazardous phthalates and chlorinated paraffins via old toys and childcare articles

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ABSTRACT

The European waste framework directive encourages reuse, refurbishment and recycling of products and materials in order to reduce plastic waste. However, thousands of chemicals are used in plastic materials. Many of these are potentially toxic, and may cause hormonal and developmental disruption in children. This includes phthalates and short chain chlorinated paraffins, which are used as plasticizers and flame-retardants. European legal frameworks regulate the amounts of these substances in toys in an effort to protect children's health and safety. Currently, limits are set to 0.1% for phthalates and 0.15% for SCCPs. Here, we have investigated levels of these compounds in toys and childcare products that were purchased prior to and after legislation on stricter exposure levels was implemented (total of 157 items, 54 and 103 new and old, respectively). We found that a larger portion of older toys and items (83.5%) contained amounts that exceed legal limits, compared to newer toys and items (29.6%). Concentrations of DEHP, BBP, DIDP, and SCCPs were significantly higher in old items, and both DEHP and DINP were found at concentrations exceeding 400,000 mg/kg in several old balls, which is approximately 40% of the weight of the toy, and 400 times above the legal limit. These findings indicate that old toys have the potential to pose a greater risk to children, and that regulations can be useful tools to protect children from exposure to toxic chemicals. We also stress that the waste framework directive, which urges reuse and repurposing of objects such as second hand items used for dress-up play, can lead to continued exposure via chemicals in older items. We conclude that movement towards circular economy threatens to expose children from legacy compounds already restricted on the market if efforts are not made to remove these items from circulation.

1. Introduction

There is an inherent tension between the European policy goals of circularity and non-toxic environment. Whereas the EU circular economy action plan (European_Commision) aims at creating more circular material flows through e.g. reuse, refurbishment and recycling, the EU chemicals strategy (European_Commision, 2020) for sustainability addresses a toxic-free environment and aims to protect citizens' health and the environment. The EU Waste Framework Directive is an important part of the implementation of the EU Circular Economy action plan. Through the principles of the waste hierarchy, waste should be prevented and reduced through re-use, recycling and recovery of materials and goods (Gharfalkar et al., 2015). There is a risk that the efforts to reduce waste allows toxic compounds to persist in goods that remain in use or are reincorporated into recycled materials (Leslie et al., 2016). This is exemplified in the purchase of second hand toys, or repurposing of older items as toys for children, when older toys may contain hazardous chemical compounds. In a recent study measuring hazardous metals in second hand toys, the author showed that these toys contained levels of several different metals that exceed current legal limits (Turner 2018), warning that old toys can present a source of hazardous materials to children.

Plastics are incorporated in a material flow where the tension between these policy goals is especially strong. While the European Strategy for Plastics in a circular economy (A new Circular Economy Action Plan, for a cleaner and more competitive Europe, 2022) contains ambitious targets for increased recycling of plastics, there is growing evidence of significant environmental and health risks associated with chemical exposure from plastics. Large numbers of chemicals associated with plastics have been identified as toxic (Groh et al., 2019; Wiesinger et al., 2021). In fact, thousands of the chemicals used in association with plastics are substances of potential concern, and there are large knowledge gaps remaining with regards to hundreds of other substances (Wiesinger et al., 2021). As humans are exposed to plastics and plastic-associated chemicals via many routes, including via food contact materials, electronics, clothing and textiles, cosmetics and toys,

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and toxic chemicals are used in many products, it becomes increasingly important to study and regulate the use and effects of these chemicals.

Children are more sensitive to many types of chemical exposures compared to adults, due to their smaller body size (and surface to volume ratio), higher metabolic rates, developing organ and hormonal systems and differences in metabolism via renal and hepatic systems (Ginsberg et al., 2004). Therefore, it is of even greater importance that their exposure routes be thoroughly investigated. Children's toys have been shown to contain toxic substances such as metals, flame-retardants, chlorinated paraffins and phthalates (Korfali et al., 2013; Ionas et al., 2014; Lioy et al., 2015; McCombie et al., 2017; Turner 2018; McGrath et al., 2021). While these chemicals may serve functional purposes in the plastic materials, many of them are also inherently dangerous to human health and more specifically, to children. Children have a high risk of exposure due to high amounts of time spent playing, and their propensity to use their mouths (Cohen Hubal, et al. 2000; Xue et al., 2007). Indeed, children's saliva has been indicated as a significant exposure pathway in leaching models (Ionas et al., 2016).

Consequences of exposure to phthalates at young developmental ages are well studied. Reports show that exposure to these compounds during early childhood can have negative health impacts, including developmental abnormalities, effect in thyroid function, increased risk of allergies and eczema, development of diabetes and obesity, adverse cognitive and behavioural outcomes, and increased cancer risks (Boas et al., 2010; Braun et al., 2013; Ejaredar et al., 2015; Legler et al., 2015; Morgenstern et al., 2017). Epidemiological studies have not only indicated the consequences of exposure to phthalates and other endocrine disrupting compounds (EDC), but also the economic impact of exposure on a societal level. Trasande et al. (2015) concluded that the burden of disease caused by EDCs costs hundreds of billions of Euros per year, within the European Union alone. Recent studies have shown that the exposure levels to phthalates that are accepted under current regulatory frameworks are not sufficiently protective to children's health. Low level exposures, which can occur via multiple routes and products, have been shown to have adverse outcomes to children´s health (Eales et al., 2022). Likewise, short-chain chlorinated paraffins (SCCPs) are considered to be substances of very high concern (SVHC) by the European Chemicals Agency (ECHA) as they are persistent, bioaccumulative and toxic (PBT). Toxicokinetic studies in rats show that short chain paraffins can cause liver and kidney toxicity, and additional studies in animal models have indicated disruptions to thyroid metabolism and neurotoxicity (Gong et al., 2018; Yang et al., 2019; Schrenk et al., 2020). While there are data limitations concerning direct human health impacts, SCCPs are wide spread in the environment and humans are exposed via multiple routes, including indoor dust (Fridén et al., 2011), food (Iino et al., 2005; Krätschmer et al., 2021) and toys (McGrath et al., 2021). In addition, SCCPs are found in humans, including in breast milk (Xia et al., 2017; Zhou et al., 2020) and blood samples (Li et al., 2017).

The chemical content of toys on the European market is controlled by different directives and regulations, including the European Toy Safety Directive (2009/48/EC). Already in 1999, the use of phthalates in toys and children's products in Europe was regulated through the 1999/815/EC decision, restricting the use of certain phthalates (DEHP, BBP, DINP, DBP, DIDP, DNOP, explanations for abbreviations can be found in Table 1) in toys and childcare articles intended to be placed in the mouth by children under three years of age (Commission Decision of 7 December 1999, 1999). Since then, restrictions related to toys and childcare articles have subsequently been tightened. In 2005, the use of the same phthalates were restricted to a maximum limit of 0.1% by mass (1000 mg/kg) of the plasticized material of toys and childcare articles (2005/84/EC). In 2009, the new Toy Safety Directive was agreed upon (2009/48/EC), which imposed stricter requirements for the use of chemicals in toys. The directive concerned "…any product or material designed or clearly intended for use in play by children of less than 14 years of age", and prohibited the use of substances that are carcinogenic, mutagenic or toxic for reproduction (CMR) in accessible parts of toys as well as a large number of allergenic fragrances in these products (The Toy Safety Directive, 2009).

Phthalates are also, since July 2020, restricted within the EU through the restricted substances list, REACH Annex XVII, regulating their use in other types of products including food contact materials, cosmetics and product packaging. Included phthalates are DEHP, BBP, DBP, DIBP, DINP, DIDP, and DNOP. These substances must not be present in more than 0.1% by mass in the plasticized materials (ECHA, 2022a,b). Within REACH, nine phthalates are placed on the Authorisation list, REACH Annex XIV (DEHP, BBP, DBP, DIBP, DIPP, DMEP, DPP, NPIPP, and DHP), which means that the substances are not allowed to be placed on the market in the EU unless a special permit has been granted (COMMISSION REGULATION (EU) 2017/999 of 13 June 2017, 2017; COMMISSION REGULATION (EU) 2020/171 of 6 February 2020, 2020; COMMISSION REGULATION (EU) 2021/2045 of 23 November 2021, 2021).

Short chain chlorinated paraffins were identified as priority substances by the Canadian EPA in 1988 owing to their detection in air, water, sediment, plankton, invertebrates and fish (Canada_EPA, 2008), and their high persistence, bioaccumulation potential and toxicity (Palm Cousins et al. 2012). Several restrictions followed in the European union (see (Glüge et al., 2016)). SCCPs were included in the REACH candidate list in 2008 (ECHA, 2008) owing to their identification as SVHCs, meeting the criteria of a PBT and very persistent, very bioaccumulative (vPvB) properties. SCCPs were also classified as suspected carcinogens (ECHA, 2021). In 2015, the legal limit for SCCPs in products to be placed on the market and used was set at 0.15% concentration by weight (European_Commision, 2015). In addition, SCCPs were identified as POP substances and included in Annex A of the Stockholm convention in 2017 (POPs_committee, 2009; COP, 2017) which requires its Parties to prohibit and/or eliminate the production, use, import and export of included chemicals.

In this study, we have utilized target chemical analyses of hazardous phthalates and short chain chlorinated paraffins to investigate whether there are any differences in the chemical content of old toys and childcare articles compared to newly purchased items. Our goal was to address whether policy frameworks are adequate to protect children from exposure to known hazards by ensuring that toys do not contain known toxicants. We also aimed to address a possible conflict in targets in environmental goals when chemicals in products act as a hindrance to circular use of materials and ultimately, a non-toxic environment. We ask whether we should encourage the reuse of old toys and items used for dress-up play, in accordance with the waste hierarchy, or whether these should be removed use and from the circular economy, i.e. via destruction.

2. Materials and methods

2.1. Collection of toys and other items

Chemical compounds were investigated in a total of 157 new and old toys, as well as in childcare articles and items used for dress-up play. These items were collected in childcare facilities and well-care clinics as well as second hand boutiques in the two largest cities in Sweden, namely Stockholm and Gothenburg. While phthalates and SCCPs are often found in polyvinylchloride (PVC) and rubber, focus here was on items that children come into contact with, and not PVC specifically. Therefore, other materials, like synthetic textiles and polyethylene (PE), were included. The toys and childcare items analyzed here were identified either as 'new' or 'old'. New items were purchased after 2014 when further restrictions in the toy directive had been implemented. Those items that were purchased as second hand stores or donated in used conditions to the public facilities were identified as 'old' and pre-date the Toy Directive (The Toy Safety Directive, 2009). Items were divided into five categories: balls, dolls, figures (mostly small animal figures), child-

Table 1
List of compounds measured. Definitions of terms can be found in the footnote[a] below the table.

Compound	Abbreviation	CAS	Reported LOQ(mg/kg)	Regulation	Classification (by regulation)	Organisational listing/agreements
diisobutyl phthalate	DIBP	84-69-5	20	REACH candidate list, Annex XIV, Annex XVII, Toy Safety Directive GHS HH	SVHC R, EDC Health hazard	SIN list TEDX list, putative EDC
dibutyl phthalate	DBP	84-74-2	20	REACH candidate list, Annex XIV, Annex XVII, *under assessment for EDC* Toy Safety Directive GHS HH GHS ENVH	SVHC R, EDC Health hazard Environmental hazard	SIN list TEDX list, putative EDC
benzylbutyl phthalate	BBP	85-68-7	20	REACH candidate list, Annex XIV, Annex XVII, Toy Safety Directive GSH HH GHS ENVH	SVHC R, EDC Health hazard Environmental hazard	SIN list TEDX list, putative EDC
di(2-ethylhexyl) phthalate	DEHP	117-81-7	50	REACH candidate list, Annex XIV, Annex XVII Toy Safety Directive GHS HH	SVHC R, EDC Health hazard	SIN list TEDX list, putative EDC
diisononyl phthalate	DINP	28,553-12-0 or 68,515-48-0	100	Toy Safety Directive, Annex XVII	*No classification*	SIN list TEDX list, putative EDC
diisodecyl phthalate	DIDP	26,761-40-0	100	Toy Safety Directive, Annex XVII, Annex III GHS ENVH	 Environmental hazard	SIN list TEDX list, putative EDC predicted HH,
di-n-octyl phthalate	DNOP	117-84-0	20	Toy Safety Directive, Annex XVII, Annex III GHS HH	 Health hazard	SIN list TEDX list, potential EDC, UNEP,
di-n-hexyl phthalate	DHP	84-75-3	20	REACH candidate list, Annex XIV, Annex III GHS HH	SVHC R Health hazard	SIN list TEDX list, putative EDC potential EDC, UNEP
di-n-pentyl phthalate	DPP	131-18-0	20	REACH candidate list, Annex XIV, Annex III GHS HH GHS ENVH	SVHC R Health hazard Environmental hazard	SIN list TEDX list, putative EDC potential EDC, EU
diisopentyl phthalate	DIPP	605-50-5	20	REACK candidate list, Annex XIV GHS HH GHS ENVH	SVHC R Health hazard Environmental hazard	SIN list
diisononyl phthalate	D-5 Mph	71,888-89-6	100	REACH candidate list, Annex XIV, Annex III GHS HH	SVHC R Health hazard	SIN list
n-pentyl-isopentyl phthalate	NPIPP	776,297-69-9	20	REACH candidate list, Annex XIV GHS HH GHS ENVH	SVHC R Health hazard Environmental hazard	SIN list
diisohexyl phthalate	DIHP	71,850-09-4	100	REACH candidate list, Annex III GHS HH	SVHC Health hazard	SIN list,
bis (2-propylheptyl) phthalate	DPHP	53,306-54-0	50	*Under assessment for EDC*	*No classification*	predicted HH
bis (2-methoxyethyl) phthalate	DMEP	117-82-8	20	REACH candidate list, Annex XIV GHS HH	SVHC R Health hazard	SIN lis, TEDX list, putative EDC
1,2-benzendicarboxyl acid di(C7–11)- alkyl ester	DHNUP	68,515-42-4	200	REACH candidate list, Annex XIV, Annex XVII, Annex III GHS HH	SVHC R Health hazard	SIN list
short chainchlorinated paraffins	SCCP	85,535-84-8	100	REACH candidate list GHS HH GHS ENVH	PBT, POP, suspected C Health hazard Environmental hazard	SIN list Stockholm convention, annex A

[a] CAS = chemical abstract services. LOQ= limit of quantification. REACH substances of very high concern (SVHC); Annex XIV = REACH authorization list; Annex XVII = REACH restriction list; Annex III (REACH) = chemicals listed here include pesticides and industrial chemicals that have been banned or severely restricted for health or environmental reasons by two or more Parties and which the Conference of the Parties has decided to subject to the PIC procedure; global harmonization system health hazard (GSH HH); global harmonization system environmental health (GHS ENVH); SIN = a list of chemicals identified by ChemSec (https://chemsec.org/business-tool/sin-list/) as SVHC according to REACH criteria, endocrine disrupting compound (EDC) listed by United Nations Environmental Programme (UNEP (IPCP 2017)) or The Endocrine Disruption Exchange (TEDX).

care items (mattresses and changing tables), and dress-up items (old clothes and handbags). Items collected in the two Swedish cities were analyzed together. A full list of the items analyzed and their assigned categories can be found in Table S1 in the supplementary materials.

2.2. Concentrations of phthalates and short chain chlorinated paraffins in toys and other items

The data presented here are a combination of results from analyses conducted on behalf of the local environmental administration agencies of each of these municipalities. Analyses handled via Eurofins Environment Testing AB, Lidköping, Sweden, at their German partner laboratory Prüfinstitut Chemische Analytik GmbH (PiCA). Eurofins is a DAkkS (German Accreditation Body) accredited testing laboratory for chemical and physico-chemical analyses of articles with direct consumer contact, including toys. The samples were cut into pieces (2–3 mm) and extracted with a solvent mix containing equal measures of ethyl acetate, cyclohexane, and acetone in an ultrasonic bath 1 h at 60 °C. Extracts were filtered through glass wool, rinsed three times with MeOH. An internal standard mix containing deuterated forms of the 17 phthalates of interest and SCCPs (see Table 1) was added to each sample. The extracts were analysed with gas chromatography, mass spectrometry electron ionization (GC/MS EI) for phthalates and GC/MS Negative Chemical Ionization (NCI) for chlorinated paraffins. Procedural blanks consisted of solvent mixtures. We utilized up-to-date Wiley/NIST databases to assist in identification. All analyses were done in accordance with good laboratory practice (GLP). At PiCA the GC-measurement systems are tuned (calibration of the mass spectrometers) after every clean up or as needed. The GC is refreshed every working day and the functioning is confirmed using an internal standard; sample quantification is performed by an external calibration and internal standards. The laboratory processing is controlled by the internal standard and by a recovery and double determination. All measurements and protocols have been confirmed in interlaboratory tests. Data from these reports, in Swedish, are publically available online or upon request (Göteborgs_stad_Miljöförvaltning, 2016; Stockholms_stad, 2017).

Chemical compounds that were included in the screening analyses were chosen based on their hazard classifications as defined by ECHA, including CMR, PBT, vPvB and EDC. Concentrations of the seventeen chemical compounds were measured. Information on abbreviations, CAS number and annotations for each compound can be found in Table 1. Measured substances were: diisobutyl phthalate (DIBP), dibutyl phthalate (DBP), benzylbutyl phthalate (BBP), di (2-ethylhexyl) phthalate (DEHP), diisononyl phthalate (DINP), diisodecyl phthalate (DIDP), di-n-octyl phthalate (DNOP), di-n-hexyl phthalate (DHP), di-n-pentyl phthalate (DPP), diisopentyl phthalate (DIPP), diisoheptyl phthalate (D-5 Mph), n-pentyl-isopentyl phthalate (NPIPP), diisohexyl phthalate (DIHP), bis (2-propylheptyl) phthalate (DPHP), di (2-metoxietyl) phthalate (DMEP), 1,2-benzendicarboxyl acid di(C7-11)- alkyl ester (DHNUP), and short chain chlorinated paraffins (SCCPs).

2.3. Statistics

Samples were collected from 2 different cities in Sweden, but we did not analyse effects of this factor, but rather addressed age of the products. Statistical analyses were performed using GraphPad Prism 7.0. In a first step, objects were assigned as either containing measurable concentrations of a tested chemical or not, and these data were compared for the 2 categories of age ('new' versus 'old') using a 2-sided Pearson Chi-square test, and then across 5 categories for type of object ('ball,' 'doll,' 'figure,' child care material,' or 'dress-up') using a non-parametric test for multiple comparisons (Dunnet C). Objects were then scored as containing concentrations above legal limits of at least one chemical, or not, and then tested again in the same way. Data were tested for normality and homogeneity of variances with the Levene test, Q-Q plots and Shapiro-Wilk tests. Non-parametric tests were chosen since the data did not fulfill assumptions for ANOVAs. In addition, the total number of chemicals detected in each object was tallied (SCCPs were counted as one).

Finally, concentrations of chemicals were compared across age and type. For these analyses, when chemicals were not detected in an item, and since it is theoretically not possible to measure the absence of the chemicals given the detection limits, we replaced 0 values with $\frac{1}{2}$ of the limit of quantification (LOQ). The non-parametric Mann Whitney U test was used to compare between the categories 'new' and 'old.'

3. Results

Analyses showed that 9 of 17 of the chemical substances investigated occurred in the toys and childcare articles. Compounds identified in the tested items include DIBP, DBP, BBP, DEHP, DINP, DIDP, DNOP, DHP, and SCCP. Additional substances that were measured but not identified in any samples are DPP, DIPP, D-5 Mph, NPIPP, DIHP, DPHP, DMEP, and DHNUP. Results of the chemical analyses for each item are displayed in Figs. 1 and 2. Table 2 indicates the number of items in each category that contained each compound, as well as the number of items in which measured levels exceed current legally accepted concentrations. A more complete description of the items included in the study and measured concentrations of phthalates and SCCPs can be found in supplementary Table S1.

102 of the 157 tested objects were found to contain at least one of the analyzed substances. The numbers of new and old items from each of the different categories are depicted in Fig. 1, indicating the number of items containing concentrations above the LOQ and concentrations above the legal limit. Of these, a significantly greater number of old items contained one or more of the substances measured here (45.164, df = 1, $p < 0.001$). The share of new items containing substances of concern was 29.6%, or 16 individual items, while 83.5% of the old toys, childcare items or dress up items did so, totaling 86 items. Our analyses also indicated that 60.1% of the old toys and childcare articles contained concentrations of target substances above the legally defined limits of the chemical substances. This is significantly more than the number of new toys and childcare articles, where 7.4% percent contained concentrations of investigated chemical substances that were above set limits (Peason's $R = 0.536$, $p < 0.001$). DEHP occurred in 64.0% and DINP in 50.4% of the old items and were the two substances that dominated our results. Old items also contained chemicals at concentrations above legal limits at a greater frequency compared to new items (Pearson's $R = 0{,}552$, $p < 0.001$). DEHP, for example, was found in 85 old items, exceeding legal limits in the following categories, shown as percent of items: childcare articles (84%), dress up items (61.5%), balls (55.5%), dolls (25%), and figures (22.7%). The corresponding percentage of items in each category found to have chemical concentrations exceeding legal limits for DINP concentrations were: childcare articles (44%), toy figures (44%) and balls (33%).

Fig. 2 shows the concentrations of the nine compounds identified in this study, indicating old versus new items, and items that exceeds legal limits. Table 2 shows the number of items in each category that contained each compound, and the number of items in which the concentration measured exceeds legal limits is indicated. In addition, most of the new items that contained a phthalate or SCCP contained only one substance (only 3 of the new items contained 2 different substances) while the majority of the old items contained multiple substances (86 items contained 2 or more substances). A red mattress cover contained all of the detected substances at levels above LOQ. A comparison of the different chemicals in new versus old items revealed that concentrations were higher in old items for BBP ($F = 50.710$, $p < 0.001$), DEHP ($F = 10.121$, $p = 0.002$), DIDP ($F = 4.468$, $p = 0.038$) and SCCPs ($F = 7.179$, $p = 0.013$). More detailed information on concentrations can be found in supplementary table S1.

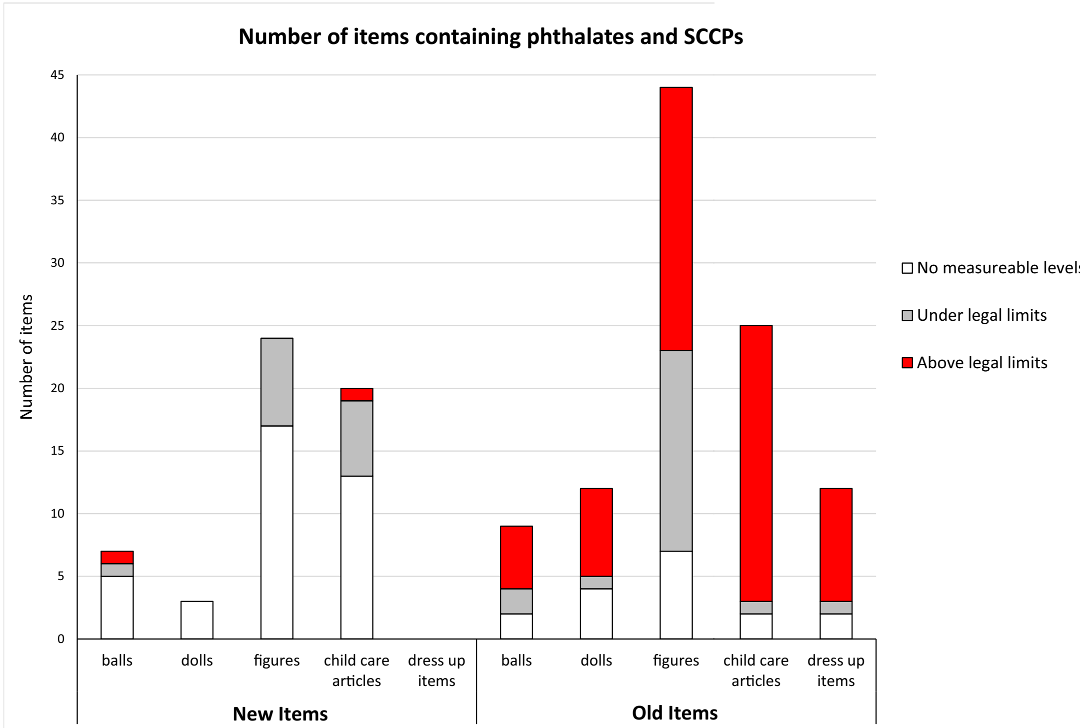

Fig. 1. The number of items containing one or more of the measured substances at levels > LOQ, indicated as below legal limits (grey) or in exceedance of legal limits (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
Number of items measured in each category that were found to contain the chemical substances listed at levels above LOQ. New items were purchased after 2014. None of the dress up items were newly purchased, so this category was excluded from the comparison here. Numbers in parentheses indicate the number of items in a category that were found to have levels of a compound exceeding current legal limits.

		New Items				Old Items					Total
	category	Balls	Dolls	Figures	Childcare articles	Balls	Dolls	Figures	Childcare articles	Dress up items	
Number of items measured		7	3	24	20	9	12	44	25	13	157
Compound	CAS										
DIBP	605-50-5	2(1)	0	4 (0)	2 (1)	2	5 (1)	21 (1)	15	8	59
DBP	84-74-2	1	0	0	0	5 (2)	8 (1)	19 (3)	23 (1)	10 (1)	66
BBP	85-68-7	0	0	0	0	0	0	3 (2)	13 (5)	1	17
DEHP	117-81-7	0	0	3	3 (2)	7 (5)	8 (4)	34 (10)	23 (21)	11 (8)	89
DINP	1,332,965-90-8	0	0	0	0	6 (3)	4 (3)	28 (16)	12 (11)	8 (7)	58
DIDP	26,761-40-0	0	0	0	0	0	1 (1)	7 (4)	17 (13)	3 (1)	28
DNOP	117-84-0	0	0	0	0	1 (1)	0	1 (1)	10 (10)	0	12
DHP	84-75-3	0	0	0	0	0	0	0	10 (5)	0	10
SCCP	85,535-84-8	1	0	0	3 (2)	0	1	6 (4)	1	4	16
Total number of items containing more than one of the substances		2	0	0	1	7	8	37	23	11	89
Shown in%		28.6	0	0	5	77.8	66.6	84.1	92	84.6	

4. Discussion

Plastics and chemicals are produced and used in vast quantities in society, to the extent that they exceed planetary boundaries and threaten the stability, health and well-being of the environment and humans (Persson et al., 2022). Indeed, this indicates the need for policies that regulate hazardous chemicals in the production, use and disposal of many different products. This study was based on chemical analyses of phthalates and short chain chlorinated paraffins and aimed to determine whether current legislation is effective in protecting children from exposure to these compounds via toys and childcare articles. Our data shows that this is in large part the case; comparing items purchased before and after the implementation of the European Toy Safety Directive in 2009 (The Toy Safety Directive, 2009), we found that the share of items containing the identified phthalates and chlorinated paraffins was significantly lower after the directive came into place. This is in line with other studies indicating that current bans in Europe have led to decreases in toxic phthalates on the market (McCombie et al., 2017). McGrath et al. (2021) found similar results regarding short chain chlorinated paraffins in toys following inclusion of these compounds in the Stockholm Convention. In addition, urinary phthalate concentrations have decreased in children and adolescents during the

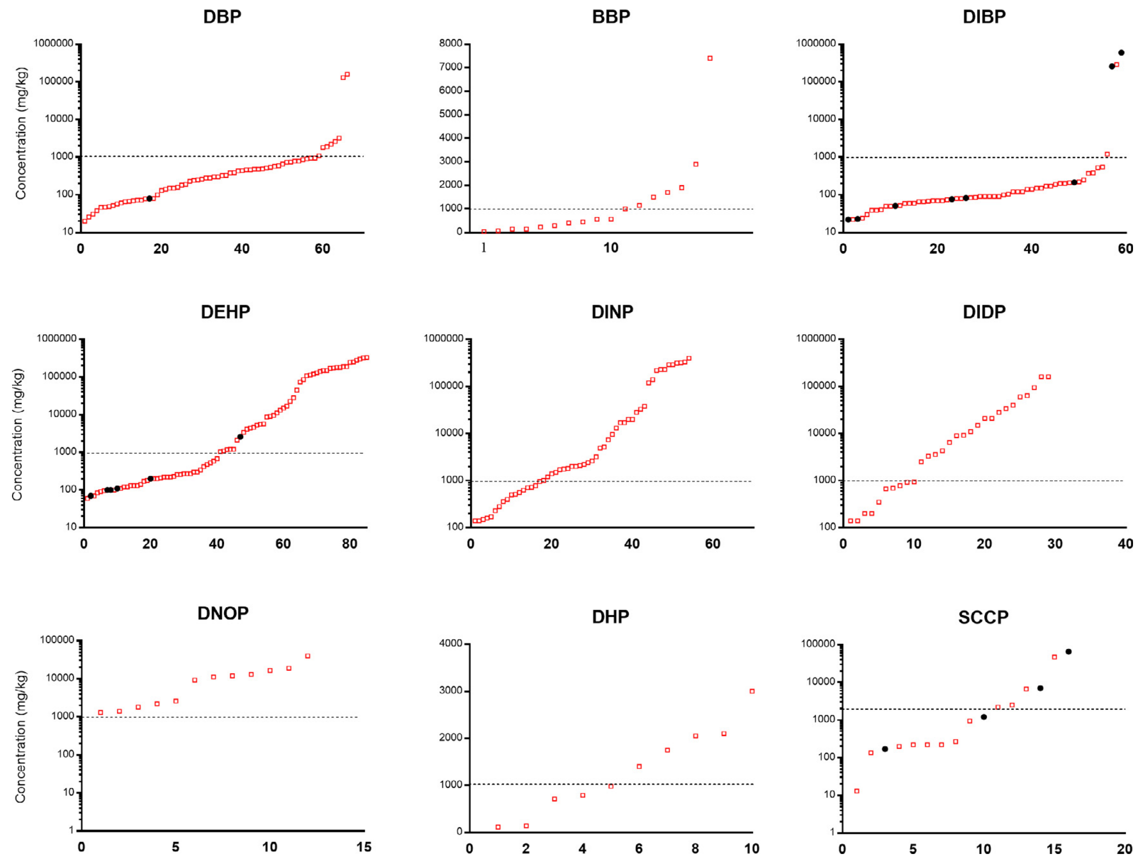

Fig. 2. Graphs depict concentrations of individual chemical compounds found in individual items, shown from lowest to highest measured concentration along the *x*-axis (numbering is arbitrary and only indicative of the total number of items containing measurable amounts of the indicated compounds). Old items are shown as open red squares (□) and new items are shown as filled black circles (●). The total number of items found to contain measurable levels of each compound are as follows: DIBP=59, DBP=66, BBP=17, DEHP=89, DINP=58, DIDP=28, DNOP=12, DHP= 10, SCCP=16. Dotted lines represent the legally permitted concentration of specific compounds in toys in Europe (1000 mg/kg for all phthalates shown, 1500 mg/kg for SCCPs). Note that both x- and y-axes differ between figures, and that y-axes are shown in log scale for all compounds, with the exception of BBP and DHP, to allow for visualization of data points over a large span of concentrations. Objects that did not contain the targeted chemicals are not shown. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

past decade (Schwedler et al., 2020). While short-chain paraffin levels have also decreased in the environment in Europe (Bogdal et al., 2017), the same is not clearly evident in human biomonitoring studies (Xu et al., 2022).

However, despite measurable declines in human exposure to phthalates (Schwedler et al., 2020; Qu et al., 2022), recent reviews of epidemiological data indicate that there is still cause for concern (Maffini et al., 2021; Eales et al., 2022). These studies report correlations between phthalate exposure (measured in blood and/or urine) and health outcomes including lower semen quality, neurodevelopmental impacts, risk of childhood asthma, and impacts on anogenital distance in boys. The authors conclude that current regulatory frameworks are evidently not protective enough, and post market analyses addressing both exposure pathways and health outcomes are needed.

Indeed, we noted an inherent risk in a second hand toy market that encourages reuse or resale of older items, as encouraged by the waste hierarchy and circular economy movements, since these toys were found to contain levels of numerous compounds that exceeded current acceptable limits. DEHP, for example, was found in a total of 83 old items, and at levels exceeding legal limits in 48 items including 55% of balls, 61% of dress up items, and −84% of child care articles. In several cases concentrations neared or exceeded 400,000 mg/kg, equal to 40% of the items by weight. This is 400 times over the legal limit today. This is also true of items repurposed for play, e.g. clothing and handbags, which contained high levels of a number of phthalates, and might be more problematic since they are not included in legislation aimed at protecting children. We found that 92% of childcare articles and 85% of dress up items contained concentrations that exceeded legal limits of at least one and up to five of the measured compounds. In fact, only 2 out of 25 ‘old’ childcare items, e.g. mattress covers, did not contain detectable amounts of DEPH and six items contained levels that exceeded today’s legal limits by more than 100x, up to 280 000 mg/kg. In addition, since the toys and childcare items included in this report were collected from state run facilities, including day care centers and baby well clinics, we argue that the items do indeed present an exposure pathway for numerous children.

Children are exposed to toxic chemicals via many pathways beyond toys and childcare articles. For example, chlorinated paraffins have been identified in baby food (Krätschmer et al., 2021), plastic sports courts and artificial turf (Cao et al., 2019), and food contact materials, which are shown to contain both chlorinated paraffins and phthalates (Wang et al., 2019). Therefore, it is important to take steps to reduce children’s exposure to these compounds also through other ways than regulations of hazardous substances in toys. A recent study demonstrated that the concentrations of phthalates in dust in preschool environments were reduced by replacing PVC flooring, with other materials (Giovanoulis et al., 2019). The stricter regulation of four phthalates (BBP, DBP, DEHP and DIBP) in electrical and electronic equipment from 2019 (in an amendment to the EU directive (RoHS 2011/62(EU)) and

in consumer products from 2020 in the European Union are likely to further reduce children´s exposure to phthalates.

Our results also show that SCCPs were present in both old and new toys and childcare articles, in total 16 out of 157 objects. Levels exceeded the legal threshold in 2 out of 4 new objects in which these compounds were identified. It is well documented that children are exposed to SCCPs via multiple routes during their lifetimes (Iino et al., 2005; Fridén et al., 2011; Krätschmer et al., 2021; McGrath et al., 2021), including at very young ages, via breast milk and potentially in utero (Xia et al., 2017; Liu et al., 2020; Zhou et al., 2020). While there is a need for increased study concerning health impacts of SCCPs, they are indicated to act as EDCs and carcinogens (Gong et al., 2018; ECHA, 2021). Numerous researchers express concern about the multitudes of low dose exposures to toxic chemicals, including EDCs like phthalates and SCCPs, that occur during early developmental stages (Gore et al., 2015; Kirchnawy et al., 2020; Ghassabian et al., 2022) and these concerns underlie such legislation as the Toy Safety Directive.

Furthermore, while we did not address manufacturing location in this study, others have shown that there is a risk for the presence of phthalates and other chemicals in goods that are produced outside of the European market (Liu et al., 2019; Praveena et al., 2021). Given that the volume of products flowing into the EU through personal orders is increasing, via e.g. direct e-commerce, and efforts to ensure consumer safety have been deficient, it is becoming necessary to increase market surveillance in the EU (Hajnal 2020). Calls are also in place to increase toy safety and production requirements in Asia, the world's largest producer and exporter of toys, as is articulated in the statement by the Association of South East Asian Nations (ASEAN) (Ismaila et al., 2020). However, until stricter regulation and surveillance is in place, consumers purchasing toys directly from outside the EU risk importing items that do not comply with EU legislation, thereby increasing risks of environmental and health impacts due to toxic chemicals (KemI, 2020).

The results of our work can also be viewed through the lens of the circular economy. Efforts to reduce waste via re-use and repurposing may enhance the problems associated with use of harmful chemicals in products, as shown in this study as well as others (Miller and Harris, 2015). In fact, recycled materials can increase phthalate exposure in children (Lee et al., 2014), and threaten to expose children from legacy compounds already restricted on the market. Toxic chemicals are also a significant hindrance to creating a circular economy of plastics (Leslie et al., 2016; de Römph and Van Calster, 2018). There are technologies available for recycling plastic materials, most commonly PE and PET but also other plastics like PVC, which may contain high levels of phthalates and SCCPs. These include incineration (sometimes referred to as energy recycling), mechanical recycling, chemical or feedstock recycling, and biological or organic recycling (Sadat-Shojai and Bakhshandeh, 2011; Shamsuyeva and Endres, 2021). While high temperature incineration can prevent significant release of legacy contaminants and additives (Wagner and Schlummer, 2020), incineration plant workers may be exposed to hazardous levels of phthalates (Lu et al., 2020), and open fires commonly used for waste management in low- and middle income countries pose a substantial risk to human health (Velis and Cook, 2021). Mechanical recycling may include a solvent phase to remove additives or to dissolve polymers and regenerate monomers for virgin-grade materials (Sherwood, 2020; Schyns and Shaver, 2021), but these practices are not implemented at scale. If the harmful chemicals and materials are not removed from the market, the positive aspects of creating more circular flows of plastics risk being outweighed by the health and environmental costs from chemical pollution (Leslie et al., 2016).

Despite this knowledge, plastic associated chemicals remain pervasive in our lives. Building on the evidence that regulations of hazardous chemicals can have intended effects on exposure, future efforts should address the use of harmful chemicals at national, regional and, importantly, global scales. Implementation of REACH policies and European chemical and waste management policies (Lee et al., 2014), together with increased transparency about chemical usage and material flows in products, could be applied to increase safety. We need to remove legacy toxicants from our material flows and homes and protect children's health.

5. Conclusions

Our study showed that phthalates and SSCPs are present in children's toys and child-care articles, but that frequency and concentrations were lower in newer items. Older items contained concentrations up to 400x more that the current legal limit, presenting a health risk and important exposure route for children, while concentrations in (most) newer items were below detection limits or legally permitted concentrations. We demonstrated that legislation can be a useful tool in removing hazardous chemicals from the market and thereby reduce exposure in children.

Declaration of Competing Interest

The authors declare that they have no conflicts of interest.

CRediT authorship contribution statement

Bethanie Carney Almroth: Conceptualization, Data curation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Resources. **Daniel Slunge:** Conceptualization, Writing – original draft, Writing – review & editing.

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